

C-axis negative magnetoresistance and upper critical field of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$.

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The out-of-plane resistance and the resistive upper critical field of BSCCO-2212 single crystals with $T_{c0} \simeq 91 - 93 \text{ K}$ have been measured in magnetic fields up to 50 T over a wide temperature range. The results are characterised by a positive linear magnetoresistance in the superconducting state and a negative linear magnetoresistance in the normal state. The zero field normal state c-axis resistance, the negative linear normal state magnetoresistance, and the divergent upper critical field $H_{c2}(T)$ are explained in the framework of the bipolaron theory of superconductivity.

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High magnetic fields have been widely used to explore the single particle spectrum of normal and superconducting metals [1]. Historically, de Haas-van Alphen effect oscillations have provided precise and detailed information on the Fermi surface and the damping of quasiparticles in Landau Fermi liquids. Such oscillations have also been studied in the vortex state of many low- T_c type-II superconductors [2] yielding information on the electronic many-body environment in the non-Fermi liquid BCS state. In the cuprates superconductors, high magnetic field studies have revealed a non-Fermi liquid temperature dependence of both ab- and c-axis resistivities [3] and a non-BCS divergent shape of the upper critical field $H_{c2}(T)$ [4–9]. These studies were performed both in relatively low- T_c cuprates [4,5,7–9] and in some high- T_c compounds in a moderate [6] (below 15 T) and high (up to 60 T) field (see Ref. [3,8] and more recent results Ref. [10]). The upper critical field was determined from the temperature dependence of c-axis resistivity with some uncertainty due to fluctuations [6]. The uncertainty was removed in the comprehensive study by Gantmakher *et al* [9] of the in-plane resistivity of high-quality $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals, which confirmed the non-BCS upper critical field observed in Ref. [4–8] strongly supporting the bipolaron theory of cuprates [11,12].

We report here on a study in pulsed magnetic fields of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, with $T_{c0} \simeq 91 - 93 \text{ K}$, which reveals new features of the c-axis transport. We observe a positive linear magnetoresistance in the flux flow (superconducting) regime and a negative linear magnetoresistance in the normal state. This allows for determination of the upper critical field, as the point of intersection of these two regimes. We have measured $H_{c2}(T)$ as a function of temperature over a wide temperature range,

$0.2 \leq T/T_{c0} \leq 1$, and find a divergent behaviour consistent with results in other materials [4–9]. We discuss this from the point of view of the bipolaron theory of superconductivity [11].

BSCCO-2212 single crystals were grown by solid state reaction [13]. Five samples with in-plane dimensions from $\simeq 85 \times 110 \mu\text{m}^2$ to $\simeq 26 \times 30 \mu\text{m}^2$ and thicknesses of $\approx 1.5 - 4.3 \mu\text{m}$ have been thoroughly studied in pulsed fields. All measurements were of the longitudinal magnetoresistance with field and current directed along the c-axis. The absence of hysteresis between the data obtained on the rising and falling sides of the pulse, characterised by very different values $\partial B/\partial t$, excludes any significant heating of our samples.

Fig.1 shows a typical measurement of the effect of magnetic field on the out-of-plane resistance of a BSCCO-2212 single crystal below a zero-field critical temperature, T_{c0} . There is a low-field regime, $R_{FF}(B, T)$, where a linear field dependence fits the experimental observations rather well, Fig.1. As has been suggested in Ref. [14] the origin of the finite c-axis resistivity below the zero field transition temperature T_{c0} might be the interplane phase slippage promoted by thermal motion of pancake vortices inside the layers. However, this mechanism does not provide the observed field dependence of the resistivity. A linear field dependence rather suggests a usual flux-flow regime. Of course, there is no such thing as flux flow resistivity for current flowing along the field direction. Nevertheless, a highly anisotropic structure of our *Bi* samples with alternating quasi-metallic and disordered non-metallic layers favors the current path with the in-plane meanders. Then there is a finite Lorentz force applied to the vortex even in the longitudinal geometry.

It is natural to attribute the high field portion of the curve in Fig.1 (assumed to be above H_{c2}) to a normal state magnetoresistance, $R_N(B, T)$, which appears to be *negative* and *linear* in B . The latter is unusual for the longitudinal transport but is also evident in other studies [3,15,16]. With these assumptions we can determine (i) the upper critical field, $H_{c2}(T)$, from the intersection of two linear approximations in Fig.1 and (ii) the zero field normal state c-axis resistance, $R_N(0, T)$, by extrapolation of the normal state linear magnetoresistance to a zero field. This procedure allows us to separate contributions originating from the normal and superconducting states and, in particular, to avoid to large extent an ambiguity due to fluctuations in the crossover region.

Referring to Fig.2, the inset shows the field dependence of BSCCO-2212 out-of-plane resistance normalised by its normal state field dependence, $R_N(B)$, thus accounting for its variation with field and temperature. The slope of the flux-flow resistance is inversely proportional to H_{c2} as $R_{FF} = R_N \times B/H_{c2}$. Indeed, H_{c2} determined from (i) the intersection of the linear fits mentioned above and (ii) that obtained from $R_{FF}(B)$ as $R_N(O, T)(\partial R_{FF}/\partial B)^{-1}$ (Fig.2), are almost identical as is seen from Fig.3 where the temperature dependence of H_{c2} is presented together with the theoretical fit using the Bose-Einstein condensation critical field [11] given by

$$H_{c2}(T) \sim (t^{-1} - t^{1/2})^{3/2} \quad (1)$$

with $t = T/T_{c0}$. $H_{c2}(T)$ shows an upward temperature dependence in agreement with the previous result based on the low field (≤ 15 T) scaling of $R(B, T)$ [6] and with independent results of other authors [4,5,7–9]. We tried (unsuccessfully) to fit the data with the pseudo-upper-critical field, $H^* \sim T^{-1} \exp(-T/T_0)$ [17] as suggested in Ref. [18] (the dashed line in Fig.3). Therefore, the model which lies behind this equation, which is based on Josephson-coupling as the origin of the anomalous $H_{c2}(T)$, is not supported by our experiment. Moreover, there is no change in the temperature dependent slope of resistivity above the superconducting transition as would be the case of superconducting domains formed well above the transition temperature. Some diamagnetism observed above the resistive $T_c(B)$ [19,18] is explained as the *normal* state Landau diamagnetism of singlet bosons [20].

The temperature dependence of the zero-field normal state resistance $R_N(0, T)$ obtained by the linear extrapolation described above, together with the theoretical curve (see below) are shown in the main panel of Fig.4 by black dots and the solid curve respectively. At low temperatures the high-field range is too short to define a linear high-field portion, so that an extrapolation to $B = 0$ to obtain directly $R_N(0, T)$ is not possible. We have found experimentally that a relation $R_N(0, T) - R_{max}(T) \propto \exp(-\alpha T)$ is valid with a constant α in the region where both the maximum value of the resistance $R_{max}(T)$ (see the arrows in Fig.1 inset) and $R_N(0, T)$ can be measured. The crosses in Fig.4 are obtained by rescaling of $R_N(0, T)$ according to this relation. The value of a dimensionless negative normal state resistance slope, $S(T) = R_N(0, T)^{-1} \partial R_N(B, T) / \partial B$, increases rapidly with falling temperature with an upturn around 50 K (the inset in Fig.4).

In what follows, we show that the unusual features of the c-axis magnetotransport, Figs. 1-4, can be broadly understood within the framework of the bipolaron theory [11,12]. As shown in Fig.3, the divergent shape of $H_{c2}(T)$ is consistent with the Bose-Einstein condensation field of charged bosons pre-formed above T_c . Within the theory the c-axis *normal*-state transport in cuprates is due

to single unpaired polarons, except at very low temperatures when unpaired carriers are frozen out [21]. Single polarons exist as excitations with the energy $k_B T^* = \Delta/2$ or larger, where Δ is the bipolaron binding energy. The edges of two polaronic bands spin-split with respect to the chemical potential (pinned at the mobility edge, $\mu \simeq E_c \equiv 0$ at low temperatures [21]) depend on the magnetic field due to a spin and orbital magnetic shifts as

$$\frac{\Delta_{\downarrow, \uparrow}}{2} = \frac{\Delta}{2} + \mu_B^* B \pm (J\sigma + \mu_B B), \quad (2)$$

where J is the exchange interaction of holes with localised copper electrons and $\sigma \equiv \langle S_i^z / S \rangle$ is an average magnetisation of copper per site. The exchange interaction leads to the spin-polarised polaron bands split by $2J\sigma$. They are further split (the last term in Eq.(2)) and shifted by the external magnetic field. Here μ_B and μ_B^* are the Bohr magnetons determined with the electron m_e and polaron m^* mass, respectively. We assume that the mobility edge is not affected by the magnetic field because bipolarons are heavier than polarons.

Assuming that $k_B T$ is less than the polaron bandwidth and noting that polarons are not degenerate at any temperature, we obtain for the polaron density

$$n_p(B, T) \sim T^{d/2} \exp[-T^*/T - \mu_B^* B / k_B T] \times \cosh[(J\sigma + \mu_B B) / k_B T], \quad (3)$$

where d is the dimensionality of the polaron energy spectrum. The exchange splitting of the polaronic bands is responsible for the negative magnetoresistance. The characteristic temperature $T_s \equiv J\sigma / k_B$ can be estimated if the magnetisation of copper ions is known. Precise measurements of magnetic neutron scattering in high-purity high- T_c single crystals of several cuprates revealed a rather low number of localised magnetic moments. The authors of these experiments noted that "what little scattering is observed, corresponds to $\simeq 3.2$ % of the Cu atoms having a spin $1/2$ " [22]. Thus, with $\sigma = 0.03$ and $J = 0.15$ eV we obtain $T_s \simeq 50$ K. It is reasonable to assume that the polaron mobility, μ_p , is field and temperature independent in the relevant region of B and T because field orbital effects are suppressed due to a heavy polaron mass. The temperature dependence of μ_p is almost absent if the scattering is dominated by a random potential [23]. As a result one obtains for the temperature dependence of the zero field normal state resistivity,

$$R_N(T) = \frac{R_0}{1 + (T/T_0)^{1/2} \exp(-T^*/T) \cosh(T_s/T)}, \quad (4)$$

where $1/R_0$ is a (low) bipolaron c-axis conductivity [23], and T_0 is a parameter depending on the ratio of the bipolaron and polaron mobilities. Here we assume $d = 1$ in agreement with the angle-resolved photoemission [24]

showing no dispersion along certain directions of the two-dimensional Brillouin zone and also with the tunnelling spectra successfully described by a one-dimensional DOS [25]. The magnetic field slope defined above is then given by

$$S(T) = \frac{\mu_B [\tanh(T_s/T) - m_e/m^*]}{k_B T} \left(\frac{R_N(T)}{R_0} - 1 \right). \quad (5)$$

As seen by reference to Fig.4, these expressions are in reasonable qualitative agreement with experiment. The solid line in Fig.4 is calculated from Eq.(4) using the following values of parameters: $R_0 = 1300 \Omega$, $T^* \simeq 170 \text{ K}$, $T_s = 53 \text{ K}$ and $T_0 = 0.046 \text{ K}$. The theoretical slope Eq.(5) is calculated with the same parameters and with $m_e/m^* = 0.022$.

Our model of c-axis magnetotransport is supported by other independent observations. While the extrapolating procedure might underestimate the magnitude of the upper critical field, its unusual temperature dependence is robust as demonstrated in the in-plane resistivity data [9]. The fact that the negative linear c-axis magnetoresistance is observed above the zero-field critical temperature tells us that this unusual phenomenon is a normal state feature rather than a signature of some fluctuations in the superconducting state. The measurements of the magnetic susceptibility [26] and the doping dependence of superconducting parameters [27] support the bipolaron origin of the normal state pseudogap T^* . The isotope effect on the normal state pseudogap observed recently [28] strongly supports its bipolaron origin as well.

In conclusion, we have measured the longitudinal out-of-plane magnetoresistance of BSCCO-2212 single crystals in magnetic fields up to 50 T. We found a quasi-linear negative magnetoresistance in the normal state and a quasi-linear positive magnetoresistance in the mixed state of BSCCO-2212. This allowed us to determine the upper critical field and to trace the zero field normal state c-axis resistance well below T_{c0} . The shape of $H_{c2}(T)$, the temperature dependence of the c-axis resistance and its negative field slope are understood within the framework of the bipolaron theory of the cuprates.

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Figure Captions

Fig.1. The magnetic field dependence of the out-of-plane resistance of BSCCO-2212 measured at 78K ($T_{c0} = 92 \text{ K}$). Linear fits to the flux-flow portion of the curve and that attributed to the normal state magnetoresistance are shown by dashed and solid lines respectively.

The inset shows the variation with field of the normal state magnetoresistance measured at different temperatures, 115, 103, 98, 90.1, 78, and 57.5 K (from the top)

normalised by the value of $R_N(0)$ (see text).

Fig.2. The temperature dependence of the flux-flow resistance slope. Inset: Field dependence of out-of-plane resistance of BSCCO-2212 normalised by its normal state value, $R_N(B)$. The selected traces are obtained (from right to the left) at 16, 20, 25, 30, 35, 45, 52.6, 57.5, 65, 70, 78, and 88.7K respectively.

Fig.3. The resistive upper critical field of BSCCO-2212 as a function of temperature obtained from the intersections of the linear extrapolations from the normal and flux-flow regimes (solid circles), and as the ratio of the extrapolated $R_N(T)$ and flux-flow resistance slope (crosses). A fit to the Bose-Einstein condensation field, Eq.(1), is shown by the solid curve, while the dashed line shows a fit to the 'pseudo-upper-critical field' of Ref. [17].

Fig.4. The zero field normal state c-axis resistance, $R_N(0)$, (main panel) and the magnetic field slope, $S(T)$, (inset) of BSCCO-2212 fitted by Eq.(4) and Eq.(5), respectively. Solid symbols in the main panel correspond to the value of $R_N(0)$ obtained by the linear extrapolation; crosses correspond to the value of $R_N(0)$ determined from R_{max} , as explained in the text.







